PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

To:

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US Department of Commerce
United States Patent and Trademark
Office, PCT
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CP2/5C24
Arlington, VA 22202
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in its capacity as elected Office

Date of mailing (day/month/year) 06 November 2000 (06.11.00)	
International application No. PCT/GB00/00577	**Applicant's or agent's file reference** SPG/P15694WO
International filing date (day/month/year) 18 February 2000 (18.02.00)	**Priority date** (day/month/year) 25 February 1999 (25.02.99)
Applicant GOODALL, Philip, Stephen et al	

1. The designated Office is hereby notified of its election made:

[X] in the demand filed with the International Preliminary Examining Authority on:

20 September 2000 (20.09.00)

[ ] in a notice effecting later election filed with the International Bureau on:

2. The election [X] was

[ ] was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO **34, chemin des Colombettes** **1211 Geneva 20, Switzerland**	Authorized officer Olivia TEFY
Facsimile No.: (41-22) 740.14.35	Telephone No.: (41-22) 338.83.38

PATENT COOPERATION TREATY

09/914320

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From the INTERNATIONAL BUREAU

To:

HARRISON GODDARD FOOTE
Belgrave Hall
Belgrave Street
Leeds LS2 8DD
ROYAUME-UNI

NOTIFICATION OF THE RECORDING OF A CHANGE

(PCT Rule 92bis.1 and Administrative Instructions, Section 422)

Date of mailing (day/month/year)	
14 December 2001 (14.12.01)	
Applicant's or agent's file reference SPG/P15694WO	**IMPORTANT NOTIFICATION**
International application No. PCT/GB00/00577	**International filing date** (day/month/year) 18 February 2000 (18.02.00)

1. The following indications appeared on record concerning:

☐ the applicant ☐ the inventor ☒ the agent ☐ the common representative

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Teleprinter No.:

2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:

☐ the person ☐ the name ☒ the address ☐ the nationality ☐ the residence

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Teleprinter No.:

3. Further observations, if necessary:

4. A copy of this notification has been sent to:

☒ the receiving Office
☐ the International Searching Authority
☐ the International Preliminary Examining Authority
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International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 7 : H01J 49/00 — A2

(11) International Publication Number: **WO 00/51160**

(43) International Publication Date: 31 August 2000 (31.08.00)

(21) International Application Number: PCT/GB00/00577

(22) International Filing Date: 18 February 2000 (18.02.00)

(30) Priority Data:
9904289.7 25 February 1999 (25.02.99) GB

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(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published
With declaration under Article 17(2)(a); without abstract; title not checked by the International Searching Authority.

(54) Title: ANALYTICAL INSTRUMENT

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ANALYTICAL INSTRUMENT

This invention relates to a novel analytical instrument, and to novel methods of measuring, *inter alia*, low concentrations of stable and radioisotopes and/or low abundance isotopes.

The determination of radionuclides at environmental levels using classical radiometric counting is well established and likely to remain the method of choice for short half-life species. However, innovations in analytical instrumentation in the last ten years have the potential to replace radiometric counting for a wide range of longer half-life species.

Elemental and isotopic analysis has advanced significantly with the introduction of plasma source mass spectrometry. A variety of plasmas have been used as ionization sources, e.g., glow discharges, microwave induced plasmas, but the inductively coupled plasma (ICP) is the most widely accepted, and *de facto*, the preferred ion source for atomic mass spectrometry. The inductively coupled plasma is compatible with solid, liquid or gaseous sample introduction and is a robust and efficient ionization source for atomic mass spectrometry.

For some potential applications of plasma mass source spectrometry, *e.g.*, environmental and biomedical monitoring of radioisotopes, current techniques may not possess the required detection limits or selectivity. Classical radiometric techniques may provide the required detection limits, but do so at the expense of protracted count times and extensive sample preparation and clean-up. For example, within a plutonium bioassay program, current radiometric methods offer detection limits of 500 μBq per litre, but require 1-2 days of sample preparation and radiometric count times of, *e.g.*, four days with α-spectrometry and up to 28 days for α-track counting. There is a requirement to develop plasma source mass spectrometry to provide enhanced selectivity and improved detection limits without sacrificing the inherent flexibility, rapidity and robustness of the technique.

The instrument of the invention is designed to measure isotopes at extremely low concentrations and isotopes of very low abundance. An example of this would be the ultra low level determination of the radionuclides. The increasing interest in the behaviour of radionuclides in the biosphere requires that new methods be developed that have detection limits equivalent to, or better than, that of the existing techniques, but combine this with superior speed and a reduced cost of analysis. Improvements in speed are essential to enable wider screening, plant and event management and to monitor illicit uses of nuclear materials. The recent OSPAR agreement has committed the UK to real reductions in levels of liquid effluent discharges. For many radionuclides, conventional radiochemical analysis will limit the ability to demonstrate that such reductions have been achieved.

To achieve the aim of improved detection limits in plasma source mass spectrometry, the factors that limit the selectivity and sensitivity of inductively coupled plasma mass spectrometry (ICP-MS) were considered. The instrumental detection limits available from ICP-MS are, in most cases, limited by the background count and not the magnitude of the analytical signal derived from the ions of interest. The background is derived broadly from three distinct sources:

1. A non-specific instrumental background.

2. Interferences from atomic or molecular ions of the same nominal mass to charge ratio, consequent upon insufficient mass spectral resolution. Examples of these "isobaric" interferences include atomic ions such as; $^{241}Am^+$, $^{241}Pu^+$; $^{90}Sr^+$, $^{90}Zr^+$; $^{55}Fe^+$, $^{55}Mn^+$; ^{40}Ca, ^{40}Ar; $^{204}Pb^+$, $^{204}Hg^+$ or molecular ions such as $^{238}U^1H^+$, $^{239}Pu^+$; $^{40}Ar^{16}O^+$, $^{56}Fe^+$,$^{40}Ar^{35}Cl^+$, $^{75}As^+$

3. Isotopes of different nominal masses but present at high relative abundances, consequent upon insufficient abundance sensitivity. For example, ^{88}Sr; ^{89}Sr, ^{90}Sr; $^{55}Fe^{+}$, $^{56}Fe^{+}$.

These observations are the key to the development of instrumentation with the superior detection limits required for determination of radionuclides at background environmental and biomedical concentrations by ICP-MS techniques.

A comparison of alternative techniques to plasma source mass spectrometry suggests that resonance ionisation mass spectrometry (RIMS) offers similar or better absolute detection limits than achieved with current generation ICP-MS instruments, *e.g.* about 4 x 10^6 atoms for ^{239}Pu. The singular advantage of RIMS over, for example, ICP-MS, is the greater isotopic selectivity derived from the laser induced ionisation process. However, the prior chemical separation, though less demanding than that required by radio-chemical methods, is nevertheless time consuming and requires specific recovery of the element, deposition onto a Ta foil and overplating with Ti. Accelerator mass spectrometry (AMS) offers absolute detection power of the order of 10^6 atoms. Selectivity is achieved through the use of high energy dissociation of molecular ions and avoidance of isobars through negative ion discrimination. Improved detection limits are obtained by high energy counting to discriminate against detector background. High abundance sensitivity is achieved by acceleration to high potentials thus minimizing the relative ion energy spread. However, AMS involves large, complex and costly instrumentation. Sample preparation is complex and time consuming, requiring preparation of the element in a pure form. For these reasons, AMS is restricted to highly specialized roles and cannot at this time be considered as a laboratory scale or general purpose instrument.

Thus, we have now developed an analytical instrument and an analytical approach that overcomes or mitigates the problems with conventionally known instruments and techniques. As a technology demonstration, this new device is based upon an ICP-MS instrument, but is equally applicable to other forms of plasma mass

spectrometry. Indeed, the range of applications includes all forms of atomic mass spectrometry and molecular mass spectrometry. This instrumentation also provides a flexible platform for spectroscopic studies of atoms and molecules to determine fundamental parameters.

Thus according to the invention, we provide an instrument comprising an Inductively Coupled Plasma Source Mass Spectrometer equipped with a multi-dimensional detector system wherein ions transmitted by the mass spectrometer are detected with high selectivity.

The instrument is provided preferably with detectors which are based upon specific detection of transmitted ions, *e.g. via.* optical spectroscopy. The device is in principle, an ICP-MS instrument operating in a multi-dimensional detection mode and including the following:

- A conventional non-specific ion detection device.

- A device based upon optical spectroscopy to provide highly selective and specific detection of ions transmitted by the mass spectrometer.

The detector device based upon optical spectroscopy provides:

- A high resolution detection system, which in conjunction with conventional mass spectrometry, is capable of resolving ions of interest from interfering molecular ions of similar nominal mass to charge ratio.

- A high resolution spectroscopy system, which in conjunction with conventional mass spectrometry, is capable of resolving ions of interest from atomic ions of similar nominal mass to charge ratio.

- A high resolution spectroscopy system, which in conjunction with conventional mass spectrometry, provides very high abundance sensitivity.

Operation of the two detection systems as a single integrated coincidence detector that provides:

- Background count rates that are orders of magnitude lower than those obtained if the individual detection systems were used as isolated, individual detectors.

The descriptive term for this approach is Inductively Coupled Plasma Mass Spectrometry Coincidence Laser Spectroscopy (ICP-MS-CLS).

Thus, according to a preferred feature of the invention, we provide an ICP- MS-CLS instrument. We especially provide an ICP-MS-CLS instrument with a conventional non-specific ion detection device and a device based on optical spectroscopy as hereinbefore defined.

The instrument of the invention supplements the universal ion counting detector with one that has a high degree of species selectivity. The use of a detector based on resonance scattering from the ions to be detected, *e.g.,* laser induced fluorescence (LIF), provides vastly improved selectivity thereby removing the problem of isobaric interferences derived from either atomic or molecular ions. Additionally, by operating the optical detector in time correlation with a second detector, background count rates can be reduced by several orders of magnitude.

The instrumentation takes advantage of improved detector technology to achieve very high spatial and temporal resolution in the optical spectroscopy. This allows coincidence detection from single photons. This capability is important in that it allows the detection of ions in which there is a high probability of trapping in a

metastable state. Ions in metastable states are transparent to the exciting laser and thus the overall photon multiplicity from these ions is low.

To allow for efficient interaction between the laser and ion beam, the ion beam must be defined accurately in space and be focussed to approximately the beam diameter of the laser. An imaging spectrometer provides an ideal solution and a sector mass spectrometer is one such device. A commercial, double focussing, sector ICP-MS provides the basic platform for development of ICP-MS-CLS.

A key feature of this instrument is the manipulation of the ion energies. To couple efficiently the energy from the laser into the ion to be detected, the optical bandwidths have to be matched. For example, an ion beam of energy of 5000 ± 2.5 eV, has a Doppler spread of about 100 MHz for an ion of mass = 240. This is in excess of the natural line width which is off the order of 15 MHz. The ion energies were manipulated by two devices. The first involves the introduction of a collision/reaction cell to act as an ion bridge between the sampler/skimmer plasma interface and the mass spectrometer. This thermalises the ions and reduces their energy spread to less than 1 eV. Additionally, it enables selective gas phase chemistry to dissociate interfering molecular ions. The second method involves acceleration of the ions to compress the optical bandwidth of the ions to be detected. For example, an ion beam of mass 240 but with a 40 000 ± 5 eV energy range has a corresponding Doppler spread of about 37 MHz. In practice, by using a collision/reaction cell, lower standing voltages, *e.g.*, 10kV, can be employed. Assuming an ion energy spread of, *e.g.*, 1 eV, at 10 kV, the Doppler spread is about 15 MHz which approximates natural line widths.

Programmed acceleration of the ions within the optical detector is important and ensures that the ions to be detected come into resonance with the exciting laser within the detection volume of the optical detector. This prevents optical trapping of the ions prior to their arrival in the detection volume of the optical detector.

The abundance sensitivity of the spectrometer can be improved by three methods:

- Where the analyte exhibits an isotope shift, the ion of interest can be brought into resonance selectively.
- Selective excitation of one hyperfine branch of an ion of interest can also be used to increase the selectivity of the mass spectrometer.
- Many ions do not exhibit an isotope shift that can be resolved optically, but acceleration of the ions induces an isotope shift by Doppler shifting the resonant frequency of the low abundant ion away from the interfering major isotope.

Where optical trapping of the ions of interest becomes significant, this may be addressed *via* the use of two-colour excitation schemes in which the metastable state is in resonance with one of the laser frequencies. To provide maximum flexibility and elemental coverage, a two-colour CW laser system was employed. A twin laser system allows a variety of excitation schemes to be used, combining single color, two color, multiphoton excitation and combinations thereof.

A multi-slit assembly was included in the instrumentation for simultaneous detection of major isotopes, to be monitored via conventional detectors, to allow isotope ratio measurements. This will also provide reference beams so that the performance of the sample introduction system and ICP ion source can be monitored continuously and optimized.

The invention will now be illustrated, but in no way limited, with reference to the following examples and the accompanying drawings, in which,

figure 1 is a schematic representation of a Coincidence Laser Spectrometer, and

figure 2 is a schematic representation of a multi-detector head including a detector based upon a Coincidence Laser Spectrometer.

Referring to figure 1, a coincidence laser spectrometer (1) comprises an optical detector (2) coupled to a voltage programmer flight tube (3), which tube is provided with a laser system (4) and a non-specific ion detector (D1). Charged beam steering optics (5) are situated adjacent to an exit port from the flight tube. The apparatus may be provided with beam dumping means (6) adjacent to spectrometer exit slits (7).

Referring to figure 2, a spectrometer assembly (8) comprises a multi-slit assembly (9) coupled to conventional ion-detectors (10 and 11) and a coincidence laser spectrometer (12) (as defined by figure 1).

Example 1

Verification of Instrument Performance – Determination of Low Abundance Isotopes, e.g. ^{10}Be

The operating characteristics of the system were established *via* an established CLS transition, *e.g.*, the Be (II) line at 313 nm which is readily accessible to a CW tunable laser. Beryllium is an important element in its own right and its high mass isotope (^{10}Be) is an important geochronometer. It is produced by nuclear spallation of oxygen by cosmic rays and reaches an equilibrium concentration in surface quartz of about 2 x 10^7 atoms per g^{-1}. An isobaric interference with ^{10}B exists, but this can be resolved in the optical detector. A reasonable measurement of ^{10}Be was made by processing of a 5g solution after removal of the major matrix elements. Other cosmogenic isotopes that might be amenable to detection include those of K, Cs, Ca, Mn, Ni, Pd, Al and the lanthanides depending on identifying suitable spectroscopic transitions.

Example 2

Determination of Pu in Urine for Bioassay Purposes.

An aliquot of urine was spiked with a Pu tracer, processed to remove the bulk of the matrix and yielded a final sample volume of 1 cm^3. This sample was analyzed by ICP-MS-CLS using a low flow sample introduction system. The isotope ratios of ^{238}Pu, ^{239}Pu, ^{240}Pu with respect to the tracer isotope were estimated. The tracer isotope was monitored on a conventional detector whilst the isotopes of interest were determined using CLS detection. Isobaric interferences from, for example, $^{238}U^+$, $^{238}U^1H^+$, $^{204}Pb^{35}Cl^+$, ^{241}Am, were resolved optically in the CLS detector. A complete chemical separation of Pu from the matrix was not required and a simple, rapid, group separation of the actinides yielded a sample suitable for analysis by ICP-MS-CLS.

Example 3

Determination of Fundamental Nuclear Parameters

Optical isotope shifts and fine structure can be used to probe nuclei for the purpose of deriving fundamental nuclear data. The ICP-MS-CLS instrumentation allows the precise measurement of optical isotope shifts using the voltage programming facilities to bring isotopes into resonance selectively with the tuneable laser operating in frequency locked mode.

CLAIMS

1. An instrument comprising an Inductively Coupled Plasma Source Mass Spectrometer equipped with a multi-dimensional detector system wherein ions transmitted by the mass spectrometer are detected with high selectivity.

2. An instrument according to claim 1 wherein the multi-dimensional detector system comprises a plurality of sub-systems which provide a unitary response.

3. An instrument according to claim 2 wherein the multi-dimensional detector system comprises two sub-systems.

4. An instrument according to claim 3 wherein the sub-systems comprise a specific detector and a non-specific detector.

5. An instrument according to claim 3 wherein the two sub-systems of the multi-dimensional detector system are correlated temporally with high resolution.

6. An instrument according to claim 5 that provides co-incidence detection of transmitted ions.

7. An instrument according to claim 4 wherein the specific detector is based on optical spectrometry.

8. An instrument according to claim 7 wherein the specific detection of the transmitted ions is *via* resonance scattering processes.

9. An instrument according to claim 8 wherein the specific detection of the transmitted ions is *via* laser induced fluorescence.

10. An instrument according to claim 8 provided with means for collecting and detecting resonantly scattered photons efficiently.

11. An instrument according to claim 8 provided with means for the detection of the resonantly scattered photons with high temporal and spatial resolution.

12. An instrument according to claim 11 wherein the detection of resonantly scattered photons is *via* an imaging photomultiplier tube.

13. An instrument according to claim 4 wherein the second detector is a non-specific ion counting device.

14. An instrument according to claim 13 wherein the non-specific ion counting device is an electron multiplier.

15. An instrument according to claim 1 provided with means for manipulating the mean ion energy thereby reducing the relative spread of the ion beams energies.

16. An instrument according to claim 15 wherein the relative spread of ion beam energies may be manipulated to compress the optical bandwidth of the transmitted ions.

17. An instrument according to claim 15 provided with means for accelerating or decelerating the transmitted ion beam to manipulate the average ion beam energy and consequently the relative spread of ion beam energies.

18. An instrument according to claim 1 wherein a front-end collision/reaction cell is used to reduce the spread of the ion beam energies and compress the optical bandwidth of the transmitted ions.

19. An instrument according to claim 1 provided with means for manipulating the ion beam energies to bring the transmitted ion beam into resonance within the detection volume of the optical detector.

20. An instrument according to claim 19 provided with means for accelerating or decelerating the ion beam.

21. An instrument according to claim 7 wherein the ion beam is accelerated to induce an optical isotope shift by Doppler shifting.

22. An instrument according to claim 1 wherein a multiple exit slit assembly is incorporated.

23. An instrument according to claim 22 wherein the dual detector assembly is mounted upon the multiple slit assembly

24. An instrument according to claim 23 wherein the dual detector assembly is mounted upon the axial exit slit.

25. An instrument according to claim 22 wherein additional non-specific ion detectors are mounted upon the multiple exit slit assembly.

26. An instrument according to claim 25 wherein additional non-specific ion detectors are mounted upon the off-axis exit slits.

27. An instrument according to claim 26 wherein the non-specific ion detectors are electron multiplier devices.

28. A method for detecting and quantifying low concentrations of stable and/or radioisotopes and/or low abundance isotopes which comprises analysing a sample in an instrument according to claim 1.

29. A method according to claim 28 wherein the species being detected is a radionuclide.

30. A method according to claim 28 wherein selectivity is enhanced by specific optical detection of transmitted ions.

31. A method according to claim 28 wherein selectivity is enhanced by specific isotopic selection via optical isotope shifts.

32. A method according to claim 28 wherein selectivity is enhanced by inducing an optical isotope shift by acceleration of the transmitted ions with subsequent Doppler shifting.

33. A method according to claim 28 wherein selectivity is enhanced by optical probing of hyperfine splitting.

34. A method according to claim 28 wherein non-specific background is reduced by co-incidence detection of transmitted ions with subsequent improved detection limit.

35. An instrument substantially as described with reference the accompanying examples and drawings.

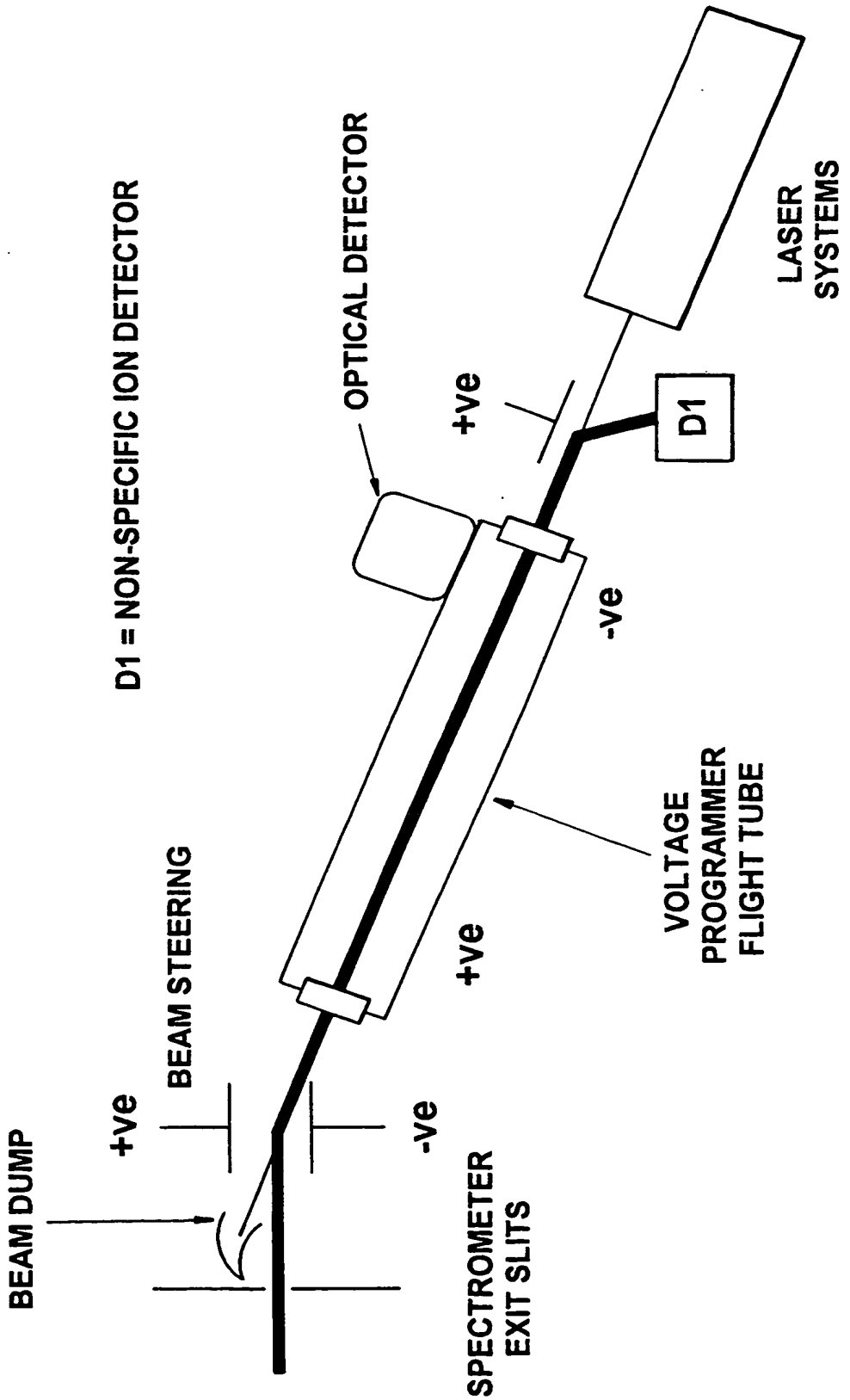
LASER SYSTEMS
D1
+ve
OPTICAL DETECTOR
D1 = NON-SPECIFIC ION DETECTOR
-ve
VOLTAGE PROGRAMMER FLIGHT TUBE
+ve
BEAM STEERING
+ve
-ve
BEAM DUMP
SPECTROMETER EXIT SLITS

Fig. 1

2/2

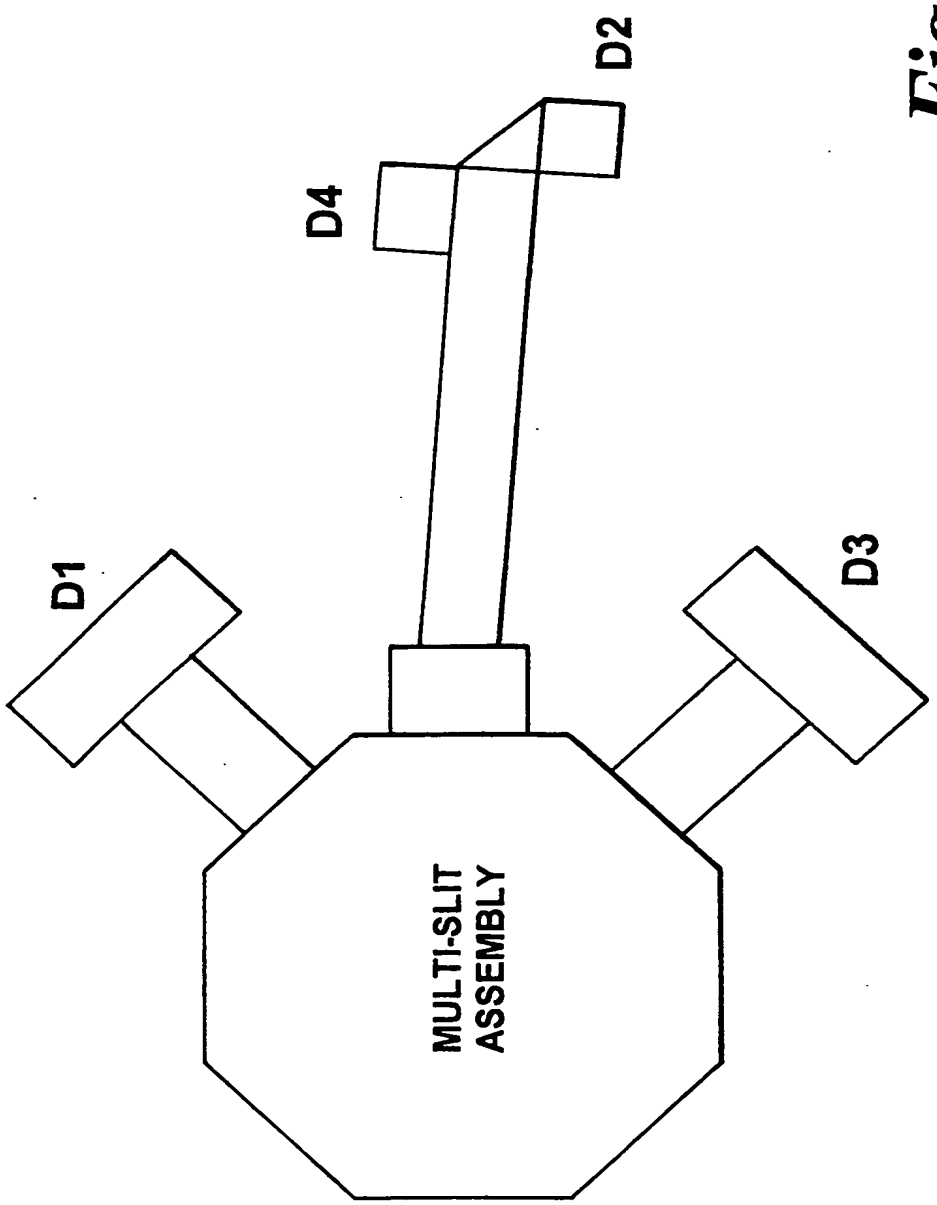

Fig. 2

D1-3 = CONVENTIONAL ION DETECTION
D4 = OPTICAL DETECTOR
N.B. A THREE SLIT ASSEMBLY HAS BEEN SHOWN FOR EXAMPLE ONLY

PATENT COOPERATION TREATY

From the INTERNATIONAL SEARCHING AUTHORITY

PCT

To:
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Tower House
Merrion Way
Leeds LS2 8PA
UNITED KINGDOM

COMMUNICATION IN CASES FOR WHICH
NO OTHER FORM IS APPLICABLE

Applicant's or agent's file reference SPG/P15694WO	Date of mailing *(day/month/year)* 23/08/2000
International application No. PCT/GB 00/00577	**REPLY DUE** See paragraph 1 below
Applicant BRITISH NUCLEAR FUELS PLC et al.	International filing date *(day/month/year)* 18/02/2000

1. ☐ REPLY DUE within _____ ~~months~~/days from the above date of mailing

   ☒ NO REPLY DUE

2. COMMUNICATION:

Please find copy of Form PCT/ISA/220 and PCT/ISA/203 with new date and new address.

Name and mailing address of the International Searching Authority European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Mildred Condron

Form PCT/ISA/224 (January 1994)

PATENT COOPERATION TREATY

From th INTERNATIONAL SEARCHING AUTHORITY

PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT OR THE DECLARATION

(PCT Rule 44.1)

To:
Harrison Goddard Foote
Tower House
Merrion Way
Leeds LS2 8PA
UNITED KINGDOM

Date of mailing *(day/month/year)*	23/08/2000
Applicant's or agent's file reference SPG/P15694WO	**FOR FURTHER ACTION** See paragraphs 1 and 4 below
International application No. PCT/GB 00/ 00577	International filing date *(day/month/year)* 18/02/2000
Applicant BRITISH NUCLEAR FUELS PLC et al.	

1. ☐ The applicant is hereby notified that the International Search Report has been established and is transmitted herewith.

Filing of amendments and statement under Article 19:
The applicant is entitled, if he so wishes, to amend the claims of the International Application (see Rule 46):

When? The time limit for filing such amendments is normally 2 months from the date of transmittal of the International Search Report; however, for more details, see the notes on the accompanying sheet.

Where? Directly to the International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland
Fascimile No.: (41-22) 740.14.35

For more detailed instructions, see the notes on the accompanying sheet.

2. ☒ The applicant is hereby notified that no International Search Report will be established and that the declaration under Article 17(2)(a) to that effect is transmitted herewith.

3. ☐ **With regard to the protest** against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:

☐ the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.

☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Further action(s):** The applicant is reminded of the following:

Shortly after **18 months** from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90*bis*.1 and 90*bis*.3, respectively, before the completion of the technical preparations for international publication.

Within **19 months** from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later).

Within **20 months** from the priority date, the applicant must perform the prescribed acts for entry into the national phase before all designated Offices which have not been elected in the demand or in a later election within 19 months from the priority date or could not be elected because they are not bound by Chapter II.

Name and mailing address of the International Searching Authority European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Mildred Condron

Form PCT/ISA/220 (July 1998)

PATENT COOPERATION TREATY

PCT

DECLARATION OF NON-ESTABLISHMENT OF INTERNATIONAL SEARCH REPORT

(PCT Article 17(2)(a), Rules 13ter.1(c) and Rule 39)

Applicant's or agent's file reference	IMPORTANT DECLARATION	Date of mailing *(day/month/year)*
SPG/P15694WO		23/08/2000
International application No.	International filing date *(day/month/year)*	(Earliest) Priority date *(day/month/year)*
PCT/GB 00/00577	18/02/2000	25/02/1999
International Patent Classification (IPC) or both national classification and IPC		H01J49/42, B01D59/44, G01N21/64, G01N21/73, H01J49/00
Applicant		
BRITISH NUCLEAR FUELS PLC et al.		

This International Searching Authority hereby declares, according to Article 17(2)(a), that **no international search report will be established** on the international application for the reasons indicated below

1. ☐ The subject matter of the international application relates to:
   - a. ☐ scientific theories.
   - b. ☐ mathematical theories
   - c. ☐ plant varieties.
   - d. ☐ animal varieties.
   - e. ☐ essentially biological processes for the production of plants and animals, other than microbiological processes and the products of such processes.
   - f. ☐ schemes, rules or methods of doing business.
   - g. ☐ schemes, rules or methods of performing purely mental acts.
   - h. ☐ schemes, rules or methods of playing games.
   - i. ☐ methods for treatment of the human body by surgery or therapy.
   - j. ☐ methods for treatment of the animal body by surgery or therapy.
   - k. ☐ diagnostic methods practised on the human or animal body.
   - l. ☐ mere presentations of information.
   - m. ☐ computer programs for which this International Searching Authority is not equipped to search prior art.

2. ☒ The failure of the following parts of the international application to comply with prescribed requirements prevents a meaningful search from being carried out:
   ☐ the description ☒ the claims ☐ the drawings

3. ☐ The failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions prevents a meaningful search from being carried out:
   - ☐ the written form has not been furnished or does not comply with the standard.
   - ☐ the computer readable form has not been furnished or does not comply with the standard.

4. Further comments: see further information PCT/ISA/203

Name and mailing address of the International Searching Authority	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Mildred Condron

Form PCT/ISA/203 (July 1998)

International Application No. PCT/GB 00/00577

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 203

Article 17(2)a)ii) PCT.
Claim 1 ill-defined. The expression "Multi-dimensional" has no generally accepted, well-defined interpretation, nor is the expression given meaning by the following dependent claims.

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure. If the application proceeds into the regional phase before the EPO, the applicant is reminded that a search may be carried out during examination before the EPO (see EPO Guideline C-VI, 8.5), should the problems which led to the Article 17(2) declaration be overcome.

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

Harrison Goddard Foote
Tower House
Merrion Way
Leeds LS2 8PA
GRANDE BRETAGNE

21.MAY 2001 15:54:06

PCT

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

	Date of mailing *(day/month/year)* 17.05.2001
Applicant's or agent's file reference SPG/P15694WO	**IMPORTANT NOTIFICATION**

International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/GB00/00577	18/02/2000	25/02/1999

Applicant
BRITISH NUCLEAR FUELS PLC et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.

2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.

3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/	Authorized officer
European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx 523656 epmu d Fax +49 89 2399 - 4465	Schuster-Kaechele, W Tel.+49 89 2399-2281

Form PCT/IPEA/416 (July 1992)

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
SPG/P15694WO		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/GB00/00577	18/02/2000	25/02/1999
International Patent Classification (IPC) or national classification and IPC		
H01J49/00		
Applicant		
BRITISH NUCLEAR FUELS PLC et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 4 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☐ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the International application
   - VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
20/09/2000	17.05.2001
Name and mailing address of the international preliminary examining authority:	Authorized officer
European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Winkelman, A Telephone No. +49 89 2399 2242

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/00577

I. Basi of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:
Description, pages:

1-9 as originally filed

Claims, No.:

1-35 as originally filed

Drawings, sheets:

1/2,2/2 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

☒ the entire international application.

☐ claims Nos. .

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

☐ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. are so unclear that no meaningful opinion could be formed (*specify*):

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.

☒ no international search report has been established for the said claims Nos. 1-35.

2. A meaningful international preliminary examination cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:

☐ the written form has not been furnished or does not comply with the standard.

☐ the computer readable form has not been furnished or does not comply with the standard.

The expression "Multi-dimensional" in claim 1 of the application has no generally accepted, well-defined interpretation, nor is the expression given meaning by the following dependent claims, the specification or the drawings.

PATENT COOPERATION TREATY

PCT

REC'D 21 MAY 2001
WIPO PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
SPG/P15694WO		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/GB00/00577	18/02/2000	25/02/1999
International Patent Classification (IPC) or national classification and IPC H01J49/00		
Applicant BRITISH NUCLEAR FUELS PLC et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 4 sheets, including this cover sheet.

   ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☐ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the international application
   - VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
20/09/2000	17.05.2001
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Winkelman, A Telephone No. +49 89 2399 2242

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/00577

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:
Description, pages:

1-9	as originally filed

Claims, No.:

1-35	as originally filed

Drawings, sheets:

1/2,2/2	as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: , which is:

   - ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
   - ☐ the language of publication of the international application (under Rule 48.3(b)).
   - ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   - ☐ contained in the international application in written form.
   - ☐ filed together with the international application in computer readable form.
   - ☐ furnished subsequently to this Authority in written form.
   - ☐ furnished subsequently to this Authority in computer readable form.
   - ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
   - ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

   - ☐ the description, pages:
   - ☐ the claims, Nos.:

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

☒ the entire international application.

☐ claims Nos. .

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

☐ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. are so unclear that no meaningful opinion could be formed (*specify*):

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.

☒ no international search report has been established for the said claims Nos. 1-35.

2. A meaningful international preliminary examination cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:

☐ the written form has not been furnished or does not comply with the standard.

☐ the computer readable form has not been furnished or does not comply with the standard.

The expression "Multi-dimensional" in claim 1 of the application has no generally accepted, well-defined interpretation, nor is the expression given meaning by the following dependent claims, the specification or the drawings.

United States Patent &
Trademark Office
Assistant Commissioner for Patents
Box PCT
20231 Washington D.C.
USA

Date of mailing : 23. 06. 00

COMMISSIONER OF PATENTS AND TRADEMARKS
BOX PCT
DOCUMENT MANAGEMENT BRANCH

RTH

PATENT COOPERATION TREATY

PCT

DECLARATION OF NON-ESTABLISHMENT OF INTERNATIONAL SEARCH REPORT

(PCT Article 17(2)(a), Rul s 13ter.1(c) and Rule 39)

Applicant's or agent's file reference	IMPORTANT DECLARATION	Date of mailing *(day/month/year)*
SPG/P15694WO		23/06/2000
International application No.	International filing date *(day/month/year)*	(Earliest) Priority date *(day/month/year)*
PCT/GB 00/ 00577	18/02/2000	25/02/1999
International Patent Classification (IPC) or both national classification and IPC		H01J49/42, B01D59/44, G01N21/64, G01N21/73, H01J49/00
Applicant		
BRITISH NUCLEAR FUELS PLC et al.		

This International Searching Authority hereby declares, according to Article 17(2)(a), that **no international search report will be established** on the international application for the reasons indicated below

1. ☐ The subject matter of the international application relates to:
   - a. ☐ scientific theories.
   - b. ☐ mathematical theories
   - c. ☐ plant varieties.
   - d. ☐ animal varieties.
   - e. ☐ essentially biological processes for the production of plants and animals, other than microbiological processes and the products of such processes.
   - f. ☐ schemes, rules or methods of doing business.
   - g. ☐ schemes, rules or methods of performing purely mental acts.
   - h. ☐ schemes, rules or methods of playing games.
   - i. ☐ methods for treatment of the human body by surgery or therapy.
   - j. ☐ methods for treatment of the animal body by surgery or therapy.
   - k. ☐ diagnostic methods practised on the human or animal body.
   - l. ☐ mere presentations of information.
   - m. ☐ computer programs for which this International Searching Authority is not equipped to search prior art.

2. ☒ The failure of the following parts of the international application to comply with prescribed requirements prevents a meaningful search from being carried out:
   ☐ the description ☒ the claims ☐ the drawings

3. ☐ The failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions prevents a meaningful search from being carried out:
   - ☐ the written form has not been furnished or does not comply with the standard.
   - ☐ the computer readable form has not been furnished or does not comply with the standard.

4. Further comments: see further information PCT/ISA/ 203

Name and mailing address of the International Searching Authority	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Roger Thomas

Form PCT/ISA/203 (July 1998)

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 203

Article 17(2)a)ii) PCT.
Claim 1 ill-defined. The expression "Multi-dimensional" has no generally accepted, well-defined interpretation, nor is the expression given meaning by the following dependent claims.

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure. If the application proceeds into the regional phase before the EPO, the applicant is reminded that a search may be carried out during examination before the EPO (see EPO Guideline C-VI, 8.5), should the problems which led to the Article 17(2) declaration be overcome.